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(54) Title: RECORDING MEDIUM					
(57) Abstract					
<p>A recording medium is provided which includes a microporous thermoplastics film having a fibrillar or reticulated structure and a contrasting underlayer. Impact on the microporous film causes it to change from an opaque to a clear state revealing the contrasting underlayer. The medium may be used to reveal impact e.g. of a golf ball on the head of a golf club to which it is attached, it may be printed in an impact or thermal printer or it may be used to detect temperature rises above a characteristic limit at which the film becomes transparent.</p>					

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RECORDING MEDIUM

This invention relates to a recording medium and to its use for recording information as a result of the 5 application of pressure (including mechanical impact) or heat.

Porous pressure - clarifiable films are disclosed in US-A-2957791 (Bechtold) and US-A-1526760 (Stevenson). 10 However, these films are formed by coating or casting onto a substrate followed by washing the partly formed film to remove solvent and any unextracted inorganic materials and are expensive and slow to make. This procedure is only applicable to materials which are used 15 in emulsion or suspension and does not lend itself to films formed by extrusion which are easier and less expensive to make.

This invention is based on the discovery that a 20 relatively thin normally opaque film of microporous plastics material in which the microporosity is developed by incorporating microparticles of a filler and then stretching can become transparent under the action of heat and or pressure to reveal a differently coloured 25 underlayer present to provide contrast or to provide

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transparent regions without any underlayer present, and that a film of this kind can record impact upon it, writing upon it by means of a stylus, typewriter, impact printer, thermal printer, pen or the like, or can reveal 5 when it has been heated to an activation temperature at which the microporous film becomes molten and its internal microporosity is destroyed.

The invention therefore provides a recording medium that 10 can be marked by heat or pressure comprising a normally opaque microporous thermoplastics film or sheet having preferably a fibrillar or reticulated structure and a contrasting layer on one face of the film, an opposite face of the film revealing the contrasting layer when it 15 becomes transparent, for example because the porosity of the film is destroyed, for example by heat or pressure. The film or sheet is present as a discrete preformed entity rather than being formed by casting onto a substrate or by a coating process.

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In one form of the recording medium the microporous film forms one external face of the medium. In this form the medium exhibits the best sensitivity to mechanical impact, and it can be used for example as a suitably 25 outlined patch for adhesion to a face of a golf club to

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record the impact of a ball on the golf club. The recording patch above described has the advantage that it can provide a permanent record of the impact of a golf ball on the face of a golf club, and that if the club is 5 repeatedly struck while the patch is present on the face of the golf club, the golfer can derive from the coincidence of successive areas of colouration an indication of how consistently he is hitting the ball. The diameter of the imprint gives an indication as to how 10 hard he has struck the ball and he can also derive this information from the degree of contrast obtained in the resulting coloured area of the patch where there is a gradation based on the force with which the ball has been struck.

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Some forms of the patch can be made relatively insensitive to appearance of colour as a result of accidental impact which facilitates their reuse in the manner aforesaid. In an alternative form of the 20 recording medium, the microporous film is protected on the face remote from the colour layer by a transparent overlayer. This form of the recording medium lends itself best to alternative applications where sensitivity to mechanical impact is less of a consideration or is 25 positively undesirable. For example, in this form the

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recording medium can be used as a writing surface and is particularly advantageous in outdoor uses where the recording medium can be made in the form where it is resistant to dirt, water or grease. Thus the recording 5 medium can be used in factory environments, or in outdoor environments where rain is a problem or even under water. If the protective overlayer is made sufficiently thick and typically above 75 micrometres in thickness then accidental impact marking is almost totally 10 prevented. The material can still be used as a recording medium, however, by printing onto the underside of the composite using an impact or thermal printer that give a mirror image print. The recording medium can be provided in the form of labels e.g. adhesive labels for marking 15 articles.

It has further been found that some forms of the recording material of the invention can be printed by a thermal printer to give a clear and precise image, and 20 they can also be printed by an impact printer of e.g. the daisy-wheel or dot-matrix type. This finds a useful application, in the provision of sheets of adhesive labels which may be printed with information relevant to a particular article before application thereto. When 25 used for making labels the microporous plastics film has

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the advantage that it is ink-receptive and can be pre-printed with non-variable information e.g. using a conventional water or oil-based printing ink or by hot foil stamping.

5

The invention also provides a recording medium that is responsive to heat and comprises a wax in or on the microporous material. A preferred further form of the invention provides wax-coated temperature indicating 10 labels in which the wax which is in or on the microporous film melts at a predetermined temperature and by entering the pores of the film and wetting it out converts the film from its opaque to its transparent state. The waxes used may include paraffin wax m.p. 50-60°C but waxes that 15 melt between 30 and 100°C may be employed e.g. blends of paraffin waxes and stearin. These waxes are conveniently applied to the microporous film as aqueous emulsions. The preparation of aqueous emulsions of waxes of melting point above 180°C is more difficult, but such 20 materials could in principle be used. Examples of these higher-melting waxes include polyamide, polyester and fluorocarbon waxes.

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The invention will now be described, by way of example only, with reference to the accompanying drawings in which;

Figure 1 is a view in section of a small portion of a
5 laminated film for attachment to the face of a golf club
to indicate the impact of a golf ball on the golf club;
and

Figure 2 is a diagram illustrating how the film of figure
1 is made.

10

In Figure 1 a laminated film for attachment to the golf club comprises a microporous top layer 1 which is normally opaque and has a white appearance but becomes transparent when it is struck by a golf ball. The layer
15 1 is preferably a highly porous very thin membrane having a fibrillar internal structure. It may, for example, be of about 60% porosity and in thickness about 12 μ m. The blind face of the microporous film 1 is attached by means of a coloured adhesive 3 to a support layer 5 of
20 inextensible but flexible material for example Melinex film. The other face of the support layer 5 has a further pressure sensitive adhesive layer 7 which has the property, when the laminated film is attached to the face of a golf club, of not competitively adhering to the face
25 of golf club even after the laminated film has recorded

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the impact of a golf ball. The pressure sensitive adhesive layer 7 is covered by a release layer 9 e.g. of siliconised paper.

5 The material used to make the normally opaque microporous plastics film is preferably polyethylene on grounds of ease of conversion into a microporous film having the required combination of properties, availability and cost. Other thermoplastics materials from which the
10 microporous film can be made include polypropylene, polyvinyl chloride, ethylene tetrafluoro- ethylene, polyvinylidene fluoride and polytetrafluoro ethylene. Porous films made from these polymers can change from an opaque to a clear state on the application of pressure or
15 heat. However, it is important to provide sufficient contrast between the opaque and clear forms of the film, and an appropriate level of force required to achieve the change of state. The factors which determine these properties are the thickness of the film, the porosity of
20 the film and the internal pore structure. In a recording medium that is to change state by melting, the melting point of the microporous film is the significant quantity and the other factors are less important. Another possible route to microporous films having the required
25 properties is precipitation by a non-solvent or "phase

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inversion" of a homogeneous solution of a polymer e.g. cellulose acetate or nitrate, the film being cast onto a belt or other moving support and then stripped therefrom. The precipitation conditions and the rate of solvent removal can be selected to give a microporous structure of the appropriate pore size.

The thickness of the microporous plastics film is preferably in the range 7 to 100 μm , preferably about 12 10 μm when it is required to change state by pressure. A thick film requires more force to become clear than a thin film, whereas a thin film may not have the required opacity and may reveal the underlying coloured layer and provide insufficient contrast. Films of less than 7 μm 15 thickness will not normally provide the required contrast whereas films whose thickness is greater than 100 μm may be too insensitive to be of practical value.

The porosity of the film is preferably 30 to 70% by 20 volume measured by the density of the film compared to the bulk density of the material from which it is made. The mean pore size is advantageously in the range from 0.1 to 2 μm and the films must have a porous and preferably a fibrillar or reticulated internal structure to 25 give the required quality of opacity. It may have a

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tortuosity of at least 1.5 and typically at least 2 measured as described in WO90/15838. The porosity in itself does not give a sufficient indication of opacity. For example, a porous film of polypropylene having a 5 porosity of 38% and a mean pore size of 0.02 μm such as is sold under the trade name CELGARD 2400 has a columnar pore structure and does not exhibit the required opacity. By "columnar" there is implied a structure in which the pores extend more or less straight through the bulk of 10 the film from one face to the other to provide continuous pathways therebetween. Films of the appropriate structure may be made by extruding a thermoplastics material containing microparticles of a hard inorganic filler so as to form a film, then longitudinally 15 stretching the film to develop the required internal structure, and preferably removing the filler. Apart from inorganic fillers there may be used organic fillers that do not melt under the processing conditions (e.g. PTFE or a rubber), polymers whose melt flow index is 20 substantially zero (e.g. ultra high molecular weight polyethylene) and polymers whose melt viscosity at the processing temperature is far greater than the carrier polymer e.g. polyethylene oxide or PVC in polyethylene. Pores may be formed, as aforesaid, using a blend of two 25 incompatible or partly incompatible polymers, one of

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which is preferably removable by extraction from the other.

The purpose of the coloured layer is to enhance the
5 contrast between the opaque and the clear forms of the
microporous film and it may comprise a coating 5 to 20 μm
thick of a coloured water-based polymer resin. The
colouring agent may be a water-soluble dye or a pigment
dissolved or dispersed in the resin. The use of a
10 water-based polymer resin rather than a solvent based
resin means that the coating becomes deposited on the
surface of the microporous film without penetrating into
its pores to any significant degree. Suitable coating
resins include vinyl acetate emulsions such as those sold
15 under the trade names EMULTEX 592 and REVACRYL 396
(Harlow Chemicals, UK). Suitable water soluble dyes are
available from Hoechst under the trade names COLANYL or
FLEXONYL. The coloured layer may also be a pre-formed
coloured film or layer attached to one face of the
20 microporous film by means of a clear adhesive. The film
may be of coloured paper or coloured plastics.

The adhesive layer can be applied to the microporous film
by reverse roller coating as indicated below. The
25 microporous film having a coloured layer on one face is a

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relatively fragile structure and it is most conveniently adhered to a support layer which may be of a polyester film, metal foil or paper etc. Where a polyester film is used, its thickness may be 25 to 100 μ m, conveniently 5 about 75 μ m. Where the recording medium is to be adhered to an article for impact recording e.g. when used in relation to golf clubs, the support layer is coated with a pressure sensitive adhesive preferably a water-based adhesive such as the vinyl acetate adhesive sold under 10 the trade name REVACRYL 396. This latter adhesive has the advantage that it can be removed easily from the face of the golf club without leaving a deposit behind. The adhesive coated face of the support layer is protected by a suitable readily removable protective material such as 15 siliconised paper, siliconised plastic or a metal foil.

When used as a writing medium the microporous plastics film (layer 1 in fig 1) is overcoated with a suitable protective layer 5 (not shown) up to 75 μ m thick, the 20 overcoating being of a transparent plastic material such as polyester film (Melinex). The provision of a protective film as aforesaid brings about a reduction in contrast and an increase in force required to convert the film from an opaque to a clear state compared to a 25 similar film without the protective layer. As stated

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above protective films greater than 75 μm in thickness may be used, e.g. for adhesive labels, but then the article may need to be mirror-image printed from its rear face.

5 The following preparative example and examples describes a method for manufacturing a porous polyethylene film and the use of that film to make recording media according to the invention.

10

PREPARATIVE EXAMPLE

A medium density polyethylene film was formed from an extrudable formulation having the following composition:

	polyethylene (Sclair 8405, Du Pont)	5.5kg
	lithium carbonate (6-8 μm particle size)	4.5kg
15	lithium stearate powder	47g

20 The above formulation was charged into a twin screw extruder and extruded at about 150°C as a rod which was then granulated. The granules were subjected to a second extrusion step in which a flat film was formed which was stretched in line to form a flat film, the flat film passing over a roller at 110°C at which stretching was initiated, stretching downstream from that roller being carried out at room temperature at a draw ratio of 14:3.

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The resulting film was then passed through a bath of dilute nitric acid to remove the filler, washed with water, further washed with organic solvents to remove the water and dried using a hot air dryer. It had a 5 thickness of 12 microns after extraction and a porosity of 38% and a pore size of about 0.17 microns. The film was generally opaque and had a milky white appearance.

EXAMPLE 1

10 A recording medium was prepared from the above mentioned porous film using apparatus as shown in figure 2 of the attached drawing which diagrammatically illustrates apparatus for coating the microporous film with a coloured adhesive layer and then attaching a support 15 layer to the adhesive layer. Microporous polyethylene film from a supply roll 10 travelled to a reverse roll coater 12 where a coating of predetermined thickness is applied from a tray containing a coloured water-based latex adhesive. The coated film passes in the direction 20 of arrow 13 through a drying tunnel 14 and thence to lamination rolls 16 where its adhesive coated face has attached to it a support of Melinex film 18 from a supply roll 20 the direction of travel being indicated by arrow 19. The lamination rolls 16 are heated to typically 25 about 50°C, and the film passes in the direction of arrow

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23 from them to chill rolls 21 and then to a take up roll 22. For application of a pressure sensitive adhesive followed by a siliconised paper protective layer the same procedure as described with reference to figure 2 is 5 followed. That is to say the recording medium and support layer is positioned in place of the supply roll 10, the exposed face of the Melinex support layer is coated with a water-based vinyl acetate adhesive (REVACRYL 396) which is selected because it can be 10 readily removed from a golf club face without leaving a sticky deposit behind. The clear adhesive is applied in the reverse roll coater 12 and the film is passed through the drying tunnel 14 and then siliconised paper from roll 20 is attached to the adhesive coated face using 15 lamination rolls 16 as before. The film is passed over the chill rolls 21 and taken up on the take up roll 22. If it is desired to use the film directly to form impact recording patches for golf clubs or other similar equipment, the film from the take up roll is passed e.g. 20 to a stamping device where it is stamped out in the required shape.

EXAMPLE 2

Instead of applying the adhesive and the siliconised 25 paper backing, the face of the recording medium at which

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the microporous plastics film 1 (fig 1) is exposed is coated with a clear adhesive using a reverse roll coater 12 as aforesaid, and the film is passed through the drying tunnel 14. A protective film of Melinex 12 um 5 thick is applied over the microporous plastics film 1 (fig 1) to provide a protective coating, again using lamination rolls 16 followed by chill rolls. The resulting protective film is taken up on the take up roll 22 and provides a writing medium. It may be cut to an 10 appropriate size e.g. into individual sheets. The film described above may also be used as a thermochromic device i.e. a device for recording a temperature excursion above a predetermined limit temperature determined by the melting temperature of the microporous 15 film, in which case the film can be cut into small test patches or continuous strips.

EXAMPLE 3

A medium density polyethylene film was formed from an 20 extrudable formulation having the following composition:

Polyethylene (Sclair 8405, DuPont)	5.5 kg
Lithium Carbonate (6 - 8 micron particle size)	4.5kg
25 Lithium Stearate Powder	47g

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The above formulation was charged into a twin screw extruder and extruded at about 150°C as a rod which was then granulated. The granules were subjected to a second extrusion step in which a film was formed which was 5 stretched in line, the flat film passing over a roller at 110°C at which stretching was initiated, stretching downstream from that roller being carried out at room temperature at a draw ratio of 14:3. The resulting film had a thickness of about 30 microns.

10

The film was then passed through a bath of dilute nitric acid to remove the filler, washed with water, further washed with organic solvents to remove the water and dried using a hot air drier. It had a porosity of 38% 15 and a pore size of about 0.2 microns. It was generally opaque and had a milky-white appearance.

A recording medium was prepared from the above film using apparatus as shown in figure 2. Micro-porous 20 polyethylene film from a supply roll (10) travels to a reverse roll coater (12) where a coating of predetermined thickness was applied from a tray containing a water based latex adhesive. The coated film passed in the direction of arrow (13) through a drying tunnel (14) and

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thence to lamination rolls (16) where its adhesive coated face had attached to it a support of coloured paper (18) from a supply roll (20) the direction of travel being indicated by arrow (19). The lamination rolls (16) were 5 heated to typically about 115°C and the film passed in the direction of arrow (23) from them to chill rolls (21) and then to a take up roll (22). The resulting film could be cut to an appropriate size for example into individual sheets. A piece of the above recording medium was passed 10 through a thermal printer and it was found that a clear and precise image could be printed onto the film.

A second piece of film was similarly passed through an impact printer (Panasonic dot matrix) which could 15 alternatively be another conventional dot matrix or daisy-wheel printer from which the inking ribbon had been removed and again it was found that a clear image could be produced.

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EXAMPLE 4

An aqueous emulsion of paraffin wax of melting point 55-60°C, supplied by Industrial Waxes Ltd of Sevenoaks, Kent, was coated onto a film prepared according to Example 3. The wax coating was applied as follows:

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The recording medium from Example 3 travelled from a supply roll (10) to a reverse roll coater (12) where a coating of pre-determined thickness was applied from a tray containing the wax emulsion. The coated film passed 5 in the direction of arrow (13) through a drying tunnel (14) and then to a take up roll (22). A label was cut from this material and placed in an oven at 55°C. After a few seconds the label changed from white to red as the wax melted and filled the pores of the porous material.

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EXAMPLE 5

A film prepared according to Example 4 was coated with a protective layer of polyester film. When this laminate 15 was placed in an oven at 55°C it changed from white to red.

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CLAIMS

1. A recording medium that can be marked by heat or pressure comprising a normally opaque microporous plastics film and a coloured layer at one face of the film, an opposite face of the film revealing the coloured layer when the porosity of the film is destroyed by heat or pressure, the film having a fibrillar or reticulated structure developed by stretching a precursor film containing microparticles of a filler and optionally removing the filler therefrom.
2. A medium according to claim 1, wherein the microporous film forms an external face of the medium.
3. A medium according to claim 1, wherein the microporous film is protected by a transparent covering layer.
4. A recording medium according to any preceding claim, wherein the colouring layer comprises a dye or pigment dispersed in a resin.
5. A medium according to claim 4, wherein the coloured layer is attached to one face of the film.

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6. A medium according to claim 4 or 5 wherein the resin is a hot melt resin.

7. A recording medium according to claim 4, wherein the 5 resin is a pressure sensitive adhesive.

8. A recording medium according to claim 7, wherein the dye and the pressure sensitive adhesive are water soluble.

10

9. A recording medium according to any of claims 1 to 3 wherein the coloured layer is a film attached to one face of the microporous film by means of a clear adhesive.

15 10. A recording medium according to any preceding claim wherein the microporous plastics film is of polyethylene.

11. A recording medium according to any of claims 1 to 9 wherein the plastics film is of polypropylene, polyvinyl 20 chloride, ethylene tetrafluoroethylene copolymer, polyvinylidene fluoride or polytetrafluoroethylene.

12. A recording medium according to any preceding claim, wherein the microporous film is 7 to 100 μ m thick.

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13. A recording medium according to any of claims 1 to 11 wherein the microporous film is 12 to 50 μm thick.

14. A recording medium according to any preceding claim,
5 wherein the microporous film has a porosity of 30 to 70%
by volume measured by density.

15. A recording medium according to claim 3, wherein the
microporous film is coated with a layer 5 to 20 μm thick
10 of a water based resin together with a dye or pigment.

16. A recording medium according to any preceding claim,
wherein the face of the microporous film to which the
coloured layer is attached carries a pressure sensitive
15 adhesive, and a support layer is adhered to the adhesive
face of the medium.

17. A recording medium according to claim 16, wherein the
support layer is a polyester film.

20

18. A recording medium according to claim 17, wherein the
thickness of the polyester film is 25 to 100 μm .

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19. A recording medium according to claim 16, 17 or 18, wherein a face of the support film remote from the microporous film is coated with an adhesive, the adhesive being further covered by a release coated paper or other removable protective layer.
5
20. A recording medium according to claim 3, wherein the transparent overlayer is of a polyester film.
- 10 21. A recording medium according to claim 3 or 20, wherein the overlayer is 5 to 75 μ m thick.
- 15 22. A recording medium according to any preceding claim having an outline conforming to the face of a golf club.
23. A recording medium according to any of claims 1 to 21, further comprising a wax in or on the microporous film that on heating to a melting temperature causes the film to become transparent.
- 20 24. Use of a recording medium as claimed in any preceding claim for recording the impact of a golf ball on the face of a golfclub.

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25. Use of a recording medium as claimed in any of claims 1 to 21 as a recording medium for receiving information by a stylus typewriter, impact printer or pen.

5 26. Use of a recording medium as claimed in any of claims 1 to 21 and 23 as a recording medium for receiving information by a thermal printer.

10 27. Use according to claim 25 or 26, wherein a mirror-image of the information to be conveyed is printed onto a normally concealed face of the recording medium.

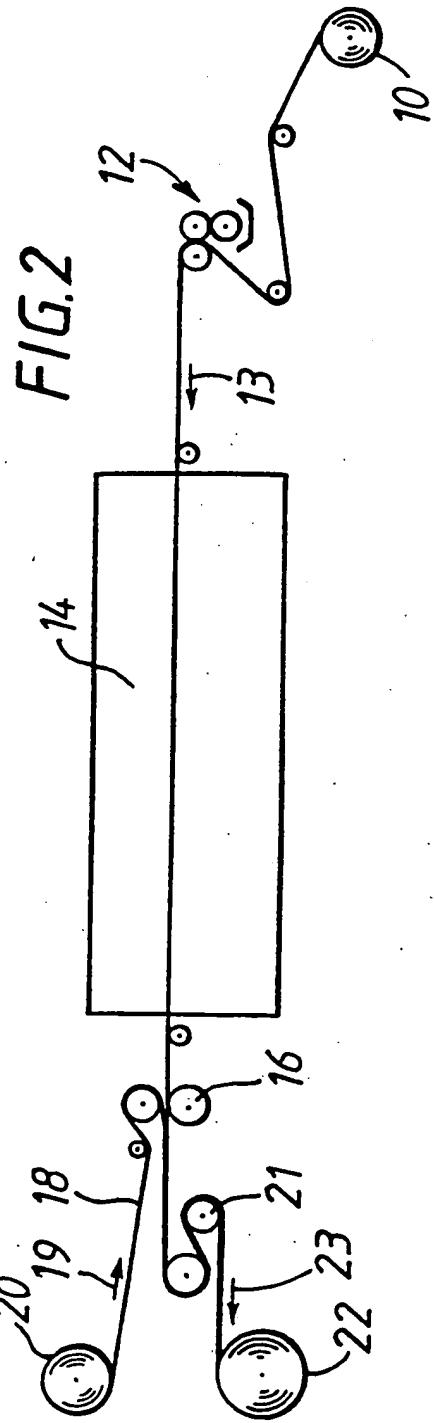
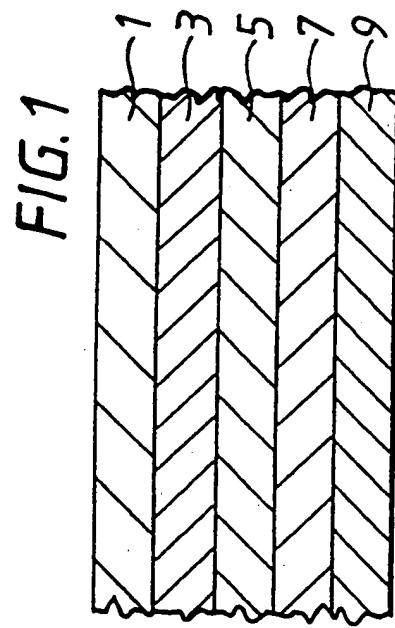
15 28. Use of a recording medium according to any of claims 1 to 21 for providing a colour change when heated to an activation temperature of the medium.

20 29. A recording medium that can be marked by heat or pressure, comprising a normally opaque microporous plastics film and a contrasting layer at one face of the film, an opposite face of the film revealing the contrasting layer when the porosity of the film is destroyed by heat or pressure, the film having a fibrillar or reticulated structure.

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30. A recording medium that can be marked by heat or pressure, comprising a normally opaque microporous plastics film and a contrasting layer at one face of the film, an opposite face of the film revealing the 5 contrasting layer when the porosity of the film is destroyed by heat or pressure, the film having a structure developed by stretching a film containing microparticles of a filler.

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INTERNATIONAL SEARCH REPORT

International Application No

PCT/GB 92/00244

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all) ⁶	
According to International Patent Classification (IPC) or to both National Classification and IPC	
Int.C1. 5 B41M5/00;	B41M5/36

II. FIELDS SEARCHED

Classification System	B41M	Minimum Documentation Searched ⁷	Classification Symbols
Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched ⁸			

III. DOCUMENTS CONSIDERED TO BE RELEVANT⁹

Category ¹⁰	Citation of Document, ¹¹ with indication, where appropriate, of the relevant passages ¹²	Relevant to Claim No. ¹³
X	DE,A,3 827 035 (BAYER AG) 15 February 1990 see the whole document ---	1-30
X	DE,A,1 778 533 (UNION CARBIDE CORPORATION) 22 July 1971 see page 26, line 1 - line 5; claim 1 ---	1-30
X	US,A,3 763 779 (S.G.PLOVAN) 9 October 1973 see column 4, line 34 - line 58 see column 7, line 8 - line 25 see column 13, line 11 - line 15 ---	1-30
A	US,A,4 318 937 (C.CEINTREY) 9 March 1982 see column 1, line 20 - line 26 ---	1-30
A	DE,B,1 301 242 (I.P. ECHEAGARAY) 14 August 1969 see the whole document ---	1-30

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IV. CERTIFICATION

Date of the Actual Completion of the International Search

1

14 APRIL 1992

Date of Mailing of this International Search Report

29 APR 1992

International Searching Authority

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Signature of Authorized Officer

BACON A.J.

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ANNEX TO THE INTERNATIONAL SEARCH REPORT
ON INTERNATIONAL PATENT APPLICATION NO. GB 9200244
SA 56320

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DE-A-1778533	22-07-71	None		
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